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A method for tractable dynamical studies of single and double shock compression

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Abstract. A new multi-scale simulation method is formulated for the study of shocked materials. The method combines molecular dynamics and the Euler equations for compressible ¤ow. Treatment of the dif£cult problem of the spontaneous formation of multiple shock waves due to material instabilities is enabled with this approach. The method allows the molecular dynamics simulation of the system under dynamical shock conditions for orders of magnitude longer time periods than is possible using the popular non-equilibrium molecular dynamics (NEMD) approach. An example calculation is given for a model potential for silicon in which a computational speedup of 10^5 is demonstrated. Results of these simulations are consistent with the recent experimental observation of an anomalously large elastic precursor on the nanosecond timescale.

INTRODUCTION

Molecular dyanmics simulations have provided valuable insight into atomic scale dynamical processes in shock waves.[1, 2, 3, 4] However, existing methods of performing these simulations are generally limited to the 10 picosecond timescale. The popular non-equilibrium molecular dynamics (NEMD) approach to atomistic simulations of shock compression involves creating a shock on one edge of a large system and allowing it to propagate until it reaches the other side. The computational work required by NEMD scales at least quadratically in the evolution time because larger systems are needed for longer simulations. When quantum mechanical methods with poor scaling of computational effort with system size are employed, this approach to shock simulations rapidly becomes impossible. Another approach that utilizes a computational cell moving at the shock speed has the same drawbacks.[5] This paper presents a method which circumvents these dif£culties by requiring simulation only of a small part of the entire system. The effects of the shock wave passing through this small piece of the system are simulated by dynamically regulating the applied stress which is obtained from a continuum theory description of the shock wave structure. Because the size of the molecular dynamics system is independent of the simulation time in this approach, the computational work required to simulate a shocked system is nearly linear in the simulation time. By circumventing the scaling problems of NEMD, molecular dyanmics simulation of shocked materials for orders of magnitude longer timescale becomes possible.

Molecular dynamics simulations have been performed that utilize a shock Hugoniot-based thermodynamic constraint for the temperature at £xed volume.[6] This approach is a thermodynamic one for a single shock wave and fails to capture the spontaneous formation of multiple shock waves and dynamical effects like long-lived metastable phases, elastic-plastic phase transitions and chemical reactions, which are ubiquitous in shocked condensed matter. The new method outlined in this paper is a method for the dynamical simulation of shock waves that solves these problems. It enables the *dynamical* simulation of shock waves in systems that have material instabilities which lead to the formation of multiple shock waves and chemical reactions that can change the speed of shock propagation with time. It is a tractable method that requires no *a priori* knowledge of the system phase diagram, metastable states,

or sound speeds.

SIMULATION OF A SINGLE SHOCK WAVE

We model the propagation of the shock wave using the 1D Euler equations for compressible ¤ow, which neglect thermal transport. These equations represent the conservation of mass, momentum, and energy respectively everywhere in the wave. Neglecting thermal transport in high temperature shocks is valid in systems where electronic mechanisms of heat conduction are not important, i.e. usually less than a few thousand K in insulators.[7] While continuum theory is not rigorously applicable at elastic shock fronts, the correct dynamics will be approximated in these special regions. We seek solutions of these equations which are steady in the frame of the shock wave moving at speed v_s. This substitution, and integration over x yields a variation of the Hugoniot relations,

$$u = v_s\left(1-\frac{\rho_0}{\rho}\right), \qquad (1)$$

$$p-p_0 = {v_s}^2\rho_0\left(1-\frac{\rho_0}{\rho}\right), \qquad (2)$$

$$e-e_0 = p_0\left(\frac{1}{\rho_0}-\frac{1}{\rho}\right)+\frac{{v_s}^2}{2}\left(1-\frac{\rho_0}{\rho}\right)^2. \qquad (3)$$

Here u is the local speed of the material in the laboratory frame (particle velocity), v is the speci£c volume, $\rho = 1/v$ is the density, e is the energy per unit mass, and p is the negative component of the stress tensor in the direction of shock propagation, $-\sigma_{xx}$. Variables with subscripts 0 are the values before the shock wave, and we have chosen $u_0 = 0$, i.e. the material is initially at rest in the laboratory frame. In the language of shock physics, Eq. 2 for the pressure is the Rayleigh line and Eq. 3 for the internal energy is the Hugoniot at constant shock velocity. These equations apply to a system which has a time-independent steady-state in the reference frame moving at the shock speed v_s.

For the molecular dynamics simulation, we employ the Lagrangian,

$$L = T\left(\{\dot{\vec{r}}_i\}\right)-V\left(\{\vec{r}_i\}\right)+\frac{1}{2}Q\dot{v}^2+\frac{1}{2}\frac{{v_s}^2}{v_0^2}(v_0-v)^2+p_0(v_0-v) \qquad (4)$$

where T and V are kinetic and potential energies per unit mass, and Q is a mass-like parameter for the simulation cell size. It can be seen that Eq. 4 implies Eq. 3 when $\dot{v}=0$ because $T+V=e$. The equation of motion for the system volume is,

$$Q\ddot{v} = \frac{\partial T}{\partial v}-\frac{\partial V}{\partial v}-p_0-\frac{{v_s}^2}{v_0^2}(v_0-v) \qquad (5)$$

which reduces to Eq. 2 when $\ddot{v}=0$. We use the scaled atomic coordinate scheme of Ref [8] to deal with the variable computational cell size. This scheme introduces a volume dependence for T and V. Strain is only allowed in the shock direction, i.e. $v_0-v = -\varepsilon_{xx}v_0$ where ε_{xx} is the uniaxial strain. The pressures in Eq. 5, including the thermal contribution, are taken to be the uniaxial x component of stresses. Computational cell dimensions transverse to the shock direction are £xed, as in NEMD simulations. This approach allows the simulation of shocks propagating in any direction which is dif£cult or impossible with NEMD.

Simulation of a single shock wave may be accomplished by dynamically varying the uniaxial strain of the system according to Eq. 5. By choosing a small representative sample of the shocked material, it is assumed that stress gradients and thermal gradients in the actual shock wave are negligible on the length scale of the sample size. While the thermal energy is assumed to be evenly distributed throughout the sample, thermal equilibrium is not required. It can be shown that the stable states of the constraint equations satisfy shock wave stability requirements. [10]

To simulate a shock to a given pressure, the initial state parameters which de£ne the MD constraint in Eq. 4 are chosen (ρ_0, p_0, e_0.) A guess for v_s is made for the constraint to take the system to the desired £nal pressure. If the £nal pressure is other than the desired one, improved guesses for v_s can be made and simulated again until the desired v_s is determined. The £nal shock pressure increases with increasing v_s. The simulation of a shock to a given particle velocity using this approach is a straightforward extension.

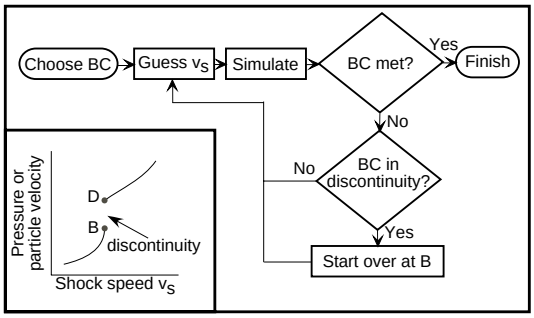

FIGURE 1. Flowchart for simulation of a shock to a chosen pressure or particle velocity boundary condition (BC). Instabilities due to regions where $\frac{d^2p}{dv^2} < 0$ along the Hugoniot can give rise to a discontinuity in the inset plot.

TREATMENT OF MULTIPLE SHOCK WAVES

The above method describes the simulation of a single stable shock wave. However, it is not always possible to shock to a given pressure or particle velocity using this technique. For example, it may not be possible to connect a straight Rayleigh line to all £nal pressures when there is a region of negative curvature in the Hugoniot, $\frac{d^2p}{dv^2} < 0$. Such regions of negative curvature are common in condensed phase materials and may be a result of phase transformations or may be the shape of a single phase Hugoniot. While a single Rayleigh line is insuf£cient to meet the pressure boundary condition in this region, two Rayleigh lines are suf£cient.

Figure 1 shows a ¤owchart that illustrates how to determine the set of Rayleigh lines that are stable and meet the boundary conditions without any *a priori* knowledge of the system. A shock wave instability exists when the boundary condition falls within a discontinuity in the set of £nal pressures as a function of shock speed, as in the inset £gure in Figure 1. The existence of such a discontinuity can be determined when suf£cient trial values of v_s have been simulated. If the boundary condition falls within the discontinuity, the entire process is repeated with point B as the initial state to £nd the shock speed that meets the boundary condition. If further instabilities are discovered that prevent the boundary condition from being met with a single shock, the process is continued.

The formation and evolution of multiple waves becomes more complicated when chemical reactions or phase transitions occur. Volume decreasing phase transformations cause the pressure at point B in Figure 1 to decrease with time. Parameterization of the *p-v* space path with Rayleigh lines is valid when the timescale of this pressure change is less than the time required for a material element to reach the £nal shocked state. This condition can be made rigorous though the so-called shock change equation and shown to hold for times longer than some timescale. [10]

APPLICATION TO SILICON

As an illustrative example, we apply the new method to an elastic-plastic transition in a model potential for silicon. Figure 2 shows shock speed as a function of particle velocity for shock waves propagating in the [011] direction in silicon described by the Stillinger-Weber potential.[11] This potential has been found to provide a qualitative representation of condensed properties of silicon. Data calculated using the NEMD method are compared with results of the new method presented in this paper. NEMD simulations were done with a computational cell of size $920\text{Å} \times 12\text{Å} \times 11\text{Å}$ unit cells (5760 atoms) for a duration of about 10-20ps. Simulations with the new method were done with a computational cell size of $19\text{Å} \times 12\text{Å} \times 11\text{Å}$ unit cells (120 atoms). Both simulations were started at 300K and zero stress. Since the NEMD simulations were limited to the 10ps timescale by computational cost, simulations with the new method were performed to calculate the Hugoniot on this 10ps timescale for comparison. The £nal particle velocity in these simulations was taken to be a point of steady state after a few ps.

Figure 2 indicates a single shock wave exists below 1.9 km/sec particle velocity. Above this particle velocity, an elastic shock wave preceeds a slower moving shock characterized by plastic deformation. Agreement between the two methods is good for all regions except for the plastic wave speed for particle velocities less than 2.1 km/sec. The wide range of values for the plastic wave speeds in NEMD simulations in this regime is due to £nite simulation cell size effects. Better agreement in this regime can be obtained by using simulation cells with larger cross

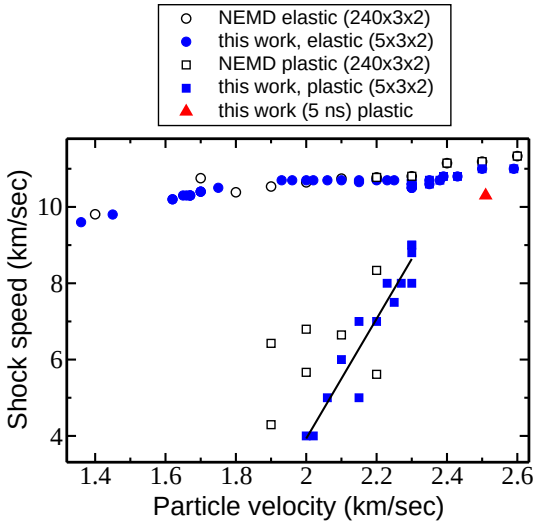

FIGURE 2. Comparison of calculated Hugoniots for the NEMD approach and the method presented in this paper for roughly 10 ps runs. Note the ability to utilize much smaller computational cell sizes with the new method. Also included is one data point for a 5 ns simulation using this work which would be prohibitive with NEMD requiring a factor of 10^5 increase in computational effort.

sectional area.

One of the primary advantages of using the method outlined in this paper is the ability to simulate for much longer times than is possible with NEMD. As an example, Figure 2 shows the result of a 5 ns simulation performed along a Rayleigh line corresponding to a shock speed of 10.3 km/sec. The uniaxially compressed elastic state required 5 ns to undergo plastic deformation. The difference in particle velocity between the 10 ps and 5 ns simulations at this shock speed is 0.8 km/sec, suggesting that the elastically compressed state is metastable with an anomalously large lifetime. This is consistent with experimental observations of shocked silicon that indicate an anomalously high pressure elastic wave exists on the nanosecond timescale.[12] In addition to the simulations performed with the Stillinger-Weber potential, we have performed more accurate tight-binding[13] 120 atom simulations using the method of this paper that also suggest an anomalously high pressure elastic wave precursor exists on the 10 ps timescale.

This simulation done with NEMD would require *more* than 5 ns simulation time due to the time required for the equilibration of the first and second wave speeds. For an $\mathcal{O}(\mathcal{N})$ method of force evaluation, the computational cost of this simulation with the NEMD method would be at least 10^5 times greater, and therefore not tractable.

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